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APPLICATION OF TRITIUM TRACER

in Soil and Plant Water Investigations

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CONTENTS

Page

Introduction 1
Principle 1
Properties of Tritium 2
Assay Methods 3
  Liquid Scintillation Counting 5
  Gas Counting 5
    G. M. Counter 5
    Ion Chamber 6
Application of Tritium 6
  Plants 7
  Soils 8
  Water Conservation and Hydrology 9
Caution and Summary 9
Literature Cited 10

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APPLICATION OF TRITIUM TRACER IN SOIL AND PLANT WATER INVESTIGATIONS

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INTRODUCTION

The use of tritium (H^3) as a tracer for ordinary hydrogen (H^1) began over 15 years ago, but the intensive application of this radioisotope has only been realized during the last 4 to 5 years with the development of improved assay equipment and techniques and the ready availability of the tracer material. The growth in the employment of tritiated compounds has been astronomical, particularly in the area of the labeling and tracing of organic metabolic compounds.

It is beyond the scope of this presentation to examine in detail the specific assay techniques and applications. The two Proceedings of the Symposia on Tritium in Tracer Applications (1957, 1958)[2] and an anonymous article in Nucleonics (1958) should prove extremely useful in regard to these topics.

The subject organization in this report will follow, in general, topics that pertain to what has been done, what is being done, and what we know can be done with tritium. It is then up to the individual investigator to appraise his own problem and decide whether he can benefit from the use of this radioisotope.

PRINCIPLE

In order that one may appreciate more fully the usefulness of tritium, a brief review will be made of the requirements of a radioisotopic tracer in general; and some of the distinct properties of tritium will be discussed. The following prerequisites can be considered to be essential in any tracer study:

- The initial concentration of the tracer must be large enough to withstand dilution during the tracing process.
- The half life of the isotope must be sufficiently long so that decay does not remove the tracer faster than it can be extracted.
- Little isotope effect should be observed; if it is present, the conclusions must be properly modified to account for this behavior.

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[2] Authors and years refer to Literature cited at end of this report.

- An adequate method of assay is necessary.

That tritium fits closely to these requirements will be seen as its properties are discussed.

PROPERTIES OF TRITIUM

Tritium is produced in nature by cosmic radiation through the following types of reactions:

$$_{7}N^{14} + {}_{o}n^{1} \longrightarrow {}_{6}C^{12} + {}_{1}H^{3}$$

$$_{8}O^{16} + {}_{1}H^{1} \longrightarrow {}_{8}O^{14} + {}_{1}H^{3}$$

As a result of the thermonuclear (hydrogen-bomb) explosions, it is estimated that the amount of tritium produced by cosmic radiation is only 10 percent of the total tritium present on the earth (Begemann, 1956).

The tritium produced by thermonuclear explosions and cosmic radiation is rapidly combined with oxygen to form water, which eventually precipitates to earth as rain or snow. The tritium content in precipitation is expressed in terms of the tritium to protium ratio (T/H), which, under normal conditions, is about 8.0×10^{-18}. It has been observed that soon after a thermonuclear explosion[3] this ratio is greatly affected, as shown in table 1.

TABLE 1.--Tritium content of rain and ocean water resulting from cosmic ray and manmade tritium

(Gilleti et al., 1958)

Source	T/H X 10^{-18} Ratio
Rain, 1953	8
Rain, 1954 (peak)	1,240
Ocean, 1953	1
Ocean, 1954	2

Tritium is obtained in the laboratory by irradiating lithium with neutrons, as follows:

$$_{3}Li^{6} + {}_{o}n^{1} \longrightarrow {}_{1}H^{3} + {}_{2}He^{4}$$

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[3] Begemann (1958) lists four thermonuclear explosions, one occurring in each of the years 1952 (Ivy), 1954 (Castle), 1955 (USSR), and 1956 (Redwing).

The radioactive material decomposes spontaneously to helium in the following manner:

$$_1H^3 \longrightarrow {}_2He^3 + \beta$$

Tritium can be obtained from the Atomic Energy Commission as a carrier-free gas at approximately 2.6 curies per cubic centimeter. The acquisition of tritiated water from commercial sources, however, is recommended instead of its preparation from tritiated gas, since special equipment and precautions are required. Tritiated water can be obtained from commercial sources at activities up to 1 curie per gram. Pure tritiated water (T_20) has an activity of approximately 3,000 curies per gram.

The radioactive properties of tritium can be best shown by comparing it in this respect with other frequently used radioisotopes (table 2).

TABLE 2.--Comparison of tritium with other radioisotopes commonly used in radiotracer studies

Isotope	Half Life	Energy of β Particle	Cost/mc	Permissible Concentration in Water
		Mev	Dollars	μc/cc
H^3	12.5 years	0.017	\$ 0.002	5×10^{-1}
C^{14}	5,600 years	.155	28.00	2×10^{-2}
P^{32}	14.7 days	1.71	1.10	5×10^{-4}

Tritium's 12.5-year half life permits the use of the tracer in long-range experiments. It is one of the cheapest radioisotopes, available at \$0.002 per mc. A 100 mc/ml sample of tritiated water costs \$30 at present commercial prices; an equivalent $C^{14}0_2$ activity $BaCO_3$ sample costs \$2,800.

Tritium in the body can be tolerated at a higher level than other radioisotopes because of its low radiation energy. The short biological half life of THO of approximately 15 days also allows for this greater latitude in the permissible concentration levels. The biological half life can be shortened considerably by increasing the intake of fluids. Revisions have been made on the permissible concentrations allowable for exposure, and the latest Handbook 69 of the (U. S.) National Bureau of Standards (1959) should be consulted.

ASSAY METHODS

The low beta energy of tritium makes assaying extremely difficult with ordinary counting equipment. It cannot be detected by end-window counters, for example, because the β-ray cannot penetrate the thinnest mica window presently available commercially. This low beta energy is not always a handicap, because it has been possible to obtain autoradiographs of very high resolutions from tritiated organic compounds. Tritiated compounds have been successfully autoradiographed in single chromosomes.

There are two distinctly different systems for the assaying of tritiated water, namely the liquid scintillation and the gas counting methods. The following listing presents the relationship:

Systems for Tritium Assay

A. Liquid Scintillation Counting (LSC)

B. Gas Counting

    1. G. M. Counter (GM)

    2. Ion Chamber (IC)

A comparison of the different counting systems is presented in table 3. The GM counting system is the most sensitive, uses the smallest sample size, and, consequently, needs the least total activity, but the preparation of the counting gas is tedious. Several papers have reported the use of tritiated water directly in the gas counting equipment (Butler, 1955; Merritt, 1958) to circumvent the sample preparation. This method of using tritiated water is hindered by a "memory effect" unless proper precautions are taken to remove the adsorbed tritiated water vapor on the counter wall between samplings.

TABLE 3.--Comparison of minimum activity of water sample detectable by different counting systems

(Guinn, 1958)

System	Volume Required	Sample Activity	Total Activity
	Gram	μc/g	μc
LSC	1.0	190 X 10^{-6}	190 X 10^{-6}
GM	.15	120 X 10^{-6}	18 X 10^{-6}
	. 15	30 X 10^{-6} (Shielding)	4.5 X 10^{-6}
IC	.37	230 X 10^{-6}	85.1 X 10^{-6}

Assaying tritiated water with the LS counting system is rapid, since the sample preparation procedures are held to a minimum. However, this requires a larger sample size or total activity and is affected by the quenching effect and limited solubility of water in the scintillation liquids.

The selection of equipment will depend primarily upon the type of research to be conducted. The ideal situation, of course, is the acquisition of both the LS and gas counting systems so that one type will complement the other.

A. Liquid Scintillation Counting (Bell and Hayes, 1958; Kinard, 1956)

The use of the LSC method has become very popular in recent years concurrent with the improvements in assay instrumentation. In this method, the tritiated water is dissolved in an organic scintillation liquid, and the phosphorescence which results from the interaction of the β particle with the liquid is detected by means of a phototube. A second scintillator liquid, a so-called wavelength shifter, is usually employed with the primary scintillator to shift the wavelength of the original emission to one for which the phototube is more sensitive.

Counting efficiencies of 5 to 10 percent for tritiated water and 15 to 30 percent for some tritiated organic compounds have been claimed by the manufacturers of the LSC instruments. Depending on whether one or two phototubes are used, and on whether a manual or automatic sample changer is present, the cost of the equipment is between $3,000 to $12,000.

The following manufacturers[4] have been supplying liquid scintillation equipment; they should be consulted for details concerning electronic components, performance data, price, etc.

1. American Tradair Corp., Long Island City, N.Y. Model Ekco N-612.
2. Baird-Atomic, Inc., Cambridge, Mass. Model 745, 745A.
3. Packard Instruments Co., Inc., LaGrange, Ill. Model Packard Tri-Carb.
4. Technical Measurement Corporation, New Haven 11, Conn. Model LP-2A.
5. Tracerlab, Inc., Waltham, Mass. Model LSC-10, 10a.

B. Gas Counting

1. G. M. Counter (Wolfgang and McKay, 1958)

In the gas counting method with the GM instrument, the tritiated water is usually reduced to TH, or to one of the lower molecular weight hydrocarbon gases giving good counting characteristics. The counting may be made either in the Geiger-Mueller or proportional region depending upon the type of gas, gas pressure, and quench mixture.

The tritiated gas, together with a suitable quenching mixture, is assayed in a specially designed counter tube. Counting efficiencies of almost 100 percent are obtainable with such a system. Anticoincidence shielding which decreases the number of background counts may be utilized in conjunction with the basic counting system to increase the sensitivity of the detection method.

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[4] In furnishing the names of manufacturers of tritium assay equipment, no attempt has been made to obtain a complete list. Therefore, no discrimination is intended against any firm whose name has been omitted; neither does the U. S. Department of Agriculture endorse the companies mentioned nor guarantee the reliability of their results.

The electronic equipment will cost anywhere from $2,000 to $5,000, the latter price for systems using anticoincidence shielding. Costs will be less if scaling equipment has been acquired previously. The high-vacuum apparatus for gas preparation will cost from $400 to $800, depending on whether some of the components are on hand.

A complete low-level counting system (Model 41, 41-1) which includes the internal gas counter, anticoincidence shielding, power supply, and scalers is commercially available[5] through the Radiation Instrument Development Laboratory, Inc., Chicago, Ill. Components of similar nature with as good performance in the system have been made by other manufacturers, but these must be assembled by the researcher.

2. Ion Chamber (Tolbert, 1956)

The equipment for preparation of the gas from water for the ion chamber is similar to that for the GM counting method, but the counting and detection units are different. The essential feature of the ion chamber is the specially insulated collection probe which is used to collect the gaseous ions produced by the β radiation from the radioactive sample. The collected charge, a function of the activity of the sample, is measured by a sensitive electrometer whose sensitivity must be in the 10^{-16} ampere range. Assaying equipment of this type will cost approximately $2,000.

Numerous manufacturers supply the various components for the ion chamber method of tritium assay. The following two concerns[6] have introduced integrated systems for lower energy detection:

1. Applied Physics Corporation, Monrovia, Calif. Model 32.
2. Nuclear-Chicago Corporation, Chicago, Ill. Dynacon System.

An important application of the ion chamber system is continuous monitoring of the tritium in the air. It also has been successfully adapted for use with the gas chromatographic analysis technique.

APPLICATION OF TRITIUM

The general areas in which tritium tracing has been or can be most profitably applied are listed below:

Areas Where Tritium can be Utilized for Tracing Water --

A. Plants

   1. Absorption of water

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[5] See footnote 4.

[6] See footnote 4.

    2. Translocation of water

    3. Distribution of water

B. Soils

    1. Movement of water (vapor and liquid) through soils

    2. Water distribution patterns in soils

    3. Interchange of water between soil and plant

C. Water conservation and hydrology

    1. Water usage by plants

    2. Ground water inventory

    3. Water circulation patterns

    4. Continental water balance

The examples presented are closely interrelated and the coverage could be easily expanded to include other types of investigation. For example, mineral uptake by plants can be studied concurrently with absorption of water by using the multiple tracer technique. A discussion of some of the investigations under the various subject headings will permit a clearer picture of tritium tracer applications.

A. Plants

In plant studies, Spalding et al. (1956) found the diffusion half-time of tritiated water in young intact bean roots to be between 1 and 2 minutes. Thimann and Samuel (1955), working with 0.5 to 4.5 mm potato tuber disks, observed that the tracer water in the disks equilibrated with the external water in approximately 20 minutes, the half-time being in the order of 1.5 minutes.

Cline (1953) reported that the plant water of bean plants grown in tritiated nutrient solution did not reach the activity of the nutrient medium in 72 hours. Incorporation of THO into the plant tissue was small (about 1 percent), the greatest quantity being present in the most actively metabolizing tissues. Biddulph and Cory (1957), in some studies of translocation in the phloem of the bean plant, measured downward tritiated water velocity of 86 cm/hr. Gage and Aronoff (1960) studied the movement of water and photosynthates by using labeled H^3 and C^{14} compounds. They concluded, from the lower recovery of THO than of solutes, that there was insufficient movement of water to fulfill the requirements of the mass flow process in the plants.

Facilities of the Plant Physiology Section of the Botany Department at Washington State University were made available to the Agricultural Research Service for investigating the applicability of tritiated water in the study of plant-soil-water relationships. Water and mineral absorption and translocation studies were initiated, the tracer water and radioisotopes of phosphorus and calcium being used. The results of a part of the experiment dealing with tritiated water are presented in table 4. This table shows the percentage of activity of the different sections of bean plants in respect to the activity of the tritiated nutrient solution at definite time intervals after the transfer of the plants to the tracer solution.

TABLE 4.--Activity of plant water in different sections of bean plants [1]

Stem Sections Above Cotyledonary Node (Inches)	THO Activity of Plant Water in Bark and Wood Related to Activity of Nutrient Solution After --					
	45 Minutes		3 Hours		72 Hours	
	Bark	Wood	Bark	Wood	Bark	Wood
	Percent	Percent	Percent	Percent	Percent	Percent
1	48.9	49.3	92.5	101.0	87.3	99.0
2	32.9	37.4	89.8	100.3	95.9	102.0
3	23.3	28.1	84.4	97.6	96.4	100.5
4	13.6	14.6	86.3	98.5	82.7	96.4
5	5.1	6.9	86.7	94.2	89.3	93.9
Leaf Sections	Petiole + Blade		Petiole + Blade		Petiole	Blade
Primary . . .	1.06		47.4		106.6	63.4
1st Trifoliate	1.65		61.6		103.0	69.0
2d Trifoliate .	1.38		65.6		98.0	62.9
3d Trifoliate .	.74		65.1		97.4	65.5
Top	.00		64.7		65.0	

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[1] Data obtained in cooperation with Dr. Orlin Biddulph, Botany Department, Washington State University, at Pullman.

The results indicate the replacement of the originally unlabeled water in the wood within 3 hours after treatment of the plant with the tracer nutrient solution. A rapid lateral movement of water from the wood to the bark is shown from the small differences in activities between the two tissues. The THO activities of the leaf blades, however, were only approximately 65 percent of the THO activity of the nutrient solution for the 72-hour experimental period. Longer time periods were not used to determine whether the leaf blade would ever attain the THO activity of the nutrient solution.

B. Soils

Few investigations have used tritiated water to study soil-water movement. The investigations of Kaufman and Orlob (1956, 2 pubs.) are the only ones in which any serious effort was made to determine the efficacy of utilizing tritium-tagged water. They found that the tracer tritium moved slower than the tracer chloride in Yolo sandy loam. The retardation is explained on the basis that there was an exchange between the THO and the originally adsorbed water. Movement of THO and chloride ion tracers was approximately the same in Hanford fine sandy loam, indicating that the retardation in materials low in clay and silt would not be significant.

Future studies in unsaturated and saturated movement of water in soils will undoubtedly involve the use of tritiated water.

C. Water Conservation and Hydrology

The measurement of tritium in our natural environment has made possible an evaluation of the whole problem of water balance on the American continent. The groups at the University of Chicago and Lamont Geological Observatory have earlier undertaken an ambitious program of determining the tritium content of rain, snow, rivers, lakes, hot springs, wells, and ocean to appraise the continental water balance, ground water inventory and storage times, surface ocean mixing rates, and worldwide water circulation patterns (Begemann, 1958; Begemann and Libby, 1958; Gilleti et al., 1958; Libby, 1959; Von Buttlar, 1959; Von Buttlar and Wendt, 1958).

The following shows in part the varied types of information obtained from the numerous measurements:

1. One-third of the rain in the Upper Mississippi Valley is ocean water; about two-thirds is reevaporated surface water.

2. Average atmospheric residence time of "manmade" tritium is 30 to 40 days.

3. Wells have been found with age of water greater than 50 years; several hot springs have water which has been stored for only a short time.

Libby (1959) suggested ways of employing tritiated water to determine underground flow patterns in a watershed, local runoff and re-evaporation rates, and ground water inventory.

CAUTION AND SUMMARY

As it is with any radioisotopic tracer, the proper precautions must be taken when using tritiated water, to prevent radiation injury to research personnel. The radiation hazard should not be ignored on the basis of the low energy. Contamination is a problem, not only from a health-physics standpoint but also in respect to the reliability of the experimental results. Because of the physical nature of the tracer material, the possibility of cross contamination within an experimental area is great.

By the use of tracer water, it is possible to answer the "yes or no" type of question; that is, for example, whether water ever moves from one part of a system to another. To get at the solution of how fast or how much water is being transported is more complicated. One must consider such things as isotopic exchange and the adsorption behavior of tritium.

The results obtained so far for the different investigations are reasonable for the system and closely correlated with other independent measurements. In some cases, tritium tracing has been the only technique available. It may be possible in the future, with improved techniques to localize the individual water molecule. This is something to which we can look forward.

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